

Measurement of Dimensionless Extinction Constant of Soot Generated Using Various Fuels

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INTRODUCTION

Even though the soot volume fraction is a key property for describing soot both in the flame and above the flame, there has been little work to verify the accuracy of measurements by light extinction techniques. Choi *et al.* [1] studied the effects of source wavelength, scattering by soot particles, light extinction by 'large' molecules and the use of different indices of refraction reported in the literature on the measurement of soot volume fraction. The experiments indicated that the measured soot volume fractions were sensitive to the absorption constant (which was calculated using the reported refractive indices). For example, at a wavelength of 632.8 nm, the absorption constant can vary by a factor of two depending on the choice of indices of refraction [1].

The focus of this paper is on the use of an independent method for characterizing soot volume fraction to assess the accuracy and to calibrate the light extinction method for soot generated using rich premixed flames. In short, the method consists of isokinetically sampling the soot at a known flow rate, measuring the mass of soot collected, and determining the density of the soot by helium pycnometry. The optical measurements can then be calibrated with the gravimetric measurements. In this manner, the dimensionless extinction constant can be determined without making assumptions regarding the optical properties of soot which can introduce significant uncertainties. The accurate measurement of the dimensionless extinction constant can improve the usefulness of the optical extinction technique.

EXPERIMENTAL APPARATUS

Figure 1 displays the experimental diagram. The burner is a 6 cm diameter water-cooled flat-flame premixed McKenna burner. A cylindrical stainless steel chimney of dimensions 8.4 cm inner diameter and 65 cm length was placed over the burner. To cool the soot/gas mixture, nitrogen was injected through two ports on the side of the steel tube. The tangential orientation of the ports led to a swirling flame with about a 3 cm diameter and 15 cm height. A 1.9 cm diameter stainless steel tripper plate was positioned 18 cm above the burner and the sampling location was about six tube diameters downstream of the tripper plate. As indicated in Figure 1, a wire mesh was positioned about 2 cm upstream of the laser beam/soot collection point to further enhance the smoke uniformity. A second screen was positioned at the top of the chimney.

The sampling probe consisted of a 13 mm stainless steel tube fitted to a filter assembly. The entrance of the probe was sharpened to a knife-edge to prevent flow stagnation near the opening. Teflon filter with a collection efficiency greater than 99% for particles larger than 0.1 μm was used [2]. The flow through the filter assembly was regulated and held constant with a mass flow controller. Since the gas flow was measured at room temperature, the temperature at the probe inlet was required to determine the actual volumetric flow of the soot/gas mixture. The temperatures were measured using a 0.25 mm diameter K-type thermocouple.

Inevitably, there were deposits of soot on the inner surface of the stainless steel probe. Soot that was used to calculate the gravimetrically-measured soot volume fraction is all of the soot that entered the probe assembly. This soot was either collected on the filter or on the inner walls of the probe. Prior to the experiment, an aluminum dish along with two fresh teflon filters were weighed. One of the filters was placed in the filter assembly shown in Fig. 1. The other filter was used to scrub the soot that was deposited on the inner walls of the probe using a plunger. The two filters and the aluminum dish were weighed again at the end of the experiment

to determine how much soot entered the probe assembly. Typically 5mg of soot was collected of which 10% was accumulated in the probe assembly.

The mass concentration of the soot, M_s , was computed from the mass of soot collected on the filter, m_s , the ratio of the ambient temperature, T_∞ , to the temperature at the probe entrance, T_p , and the total volume, V , of gas sampled based on the ambient temperature:

$$M_s = \frac{m_s}{V} \frac{T_\infty}{T_p} \quad (1)$$

The soot volume fraction based on gravimetric technique, f_v , is equal to M_s divided by the density of soot. The estimated uncertainty (one standard deviation) in m_s , V and T_p are 0.03 mg, 0.08 L and 10 K, respectively. The optical measurements were performed using a 10 mW 632.8 nm He-Ne laser and a biased photodiode with a neutral density and HeNe interference filters. For soot volume fraction measurements, the following Bouguer's Law is used:

$$\frac{I}{I_0} = \exp\left(-\frac{K_a(1 + \alpha_{sa})f_{va}L}{\lambda}\right) = \exp\left(-\frac{K_e f_{va}L}{\lambda}\right) \quad (2)$$

where L is the path length, I is the transmitted laser intensity, I_0 is the incident laser intensity, f_{va} is soot volume fraction based on optical measurements and α_{sa} is the scattering to absorption ratio. For soot particles of small optical dimension, the dimensionless absorption constant, K_a , is computed from the following formula obtained from Mie theory in the limit of small particle size using the dispersion relationship of Dalzell and Sarofim [4]:

$$K_a = \frac{36\pi n_\lambda k_\lambda}{(n_\lambda^2 - k_\lambda^2 + 2)^2 + 4n_\lambda^2 k_\lambda^2} = 4.9 \quad (3)$$

It is common practice in the combustion and fire community to set α_{sa} (scattering to absorption ratio) equal to zero to determine the soot volume fraction [1,5]. However, the assumption that the scattering to absorption ratio α_{sa} is negligible is only valid for very small aggregates with optical sizes $2\pi R_g/\lambda$ less than 0.7 (where R_g is the radius of gyration of the agglomerate [6]). With this assumption, the Rayleigh-limit solution of the soot extinction constant can be used (Eq. 3). This practice may be valid when considering the primary particle diameter (which are typically 20 to 50 nm compared to source wavelength of 632.8 nm) as the dimension of interest. However, soot particles are aggregates composed of hundreds of primary particles and the average aggregate dimension can be of the same order of magnitude as the extinction source wavelength. Under this condition, the Rayleigh-limit assumption is no longer valid and using Eq. 4 to calculate the dimensionless extinction constant will result in large uncertainties. For example, the work of Köylü and Faeth [7], indicates that the scattering to absorption ratio for soot created in the overfire region of diffusion flames can be as high as 22 to 41% (using $\lambda = 514.5$ nm).

EXTINCTION CONSTANT MEASUREMENTS

Experiments were performed for rich acetylene/air premixed flames with an equivalence ratio of 2.5. For each experiment, extinction measurements were performed in conjunction with the gravimetric soot sampling experiments (see fig. 2). One minute of laser transmission was taken prior to flame ignition. Light extinction pathlength was maintained at 25 mm by inserting the sliding glasstubes within the chimney. These tubes were

purged with slow nitrogen flow to prevent soot accumulation. Although the soot volume fraction distribution within the pathlength fluctuates, experiments performed using similar chimney arrangement indicated that the soot volume fractions attained a uniform profile after only a few seconds of averaging [8]. The sampling probe was inserted one minute after ignition to avoid the initial transient soot accumulation. The signal transmitted through the soot dispersion displayed some fluctuations (caused by small changes in the path length), however, the averaged values remained nearly constant throughout the duration of the experiment. The flame was extinguished after 5 minutes of soot sampling and the probe was immediately removed from the chimney.

For all cases, the optically determined soot volume fractions were approximately twice as large as the gravimetric measurements. Another choice of refractive index could lead to better or poorer agreement between the optical and gravimetric measurements. For example, the calculated absorption constant using the various indices of refraction can vary by a factor of two [1]. In addition to the uncertainty in the optical properties, replacing K_e with K_a neglects light scattering. This can result in as much as 25% underestimate of K_e and 33% overestimate of the soot volume fraction. However, the gravimetrically determined soot volume fraction is calculated to be accurate to within $\pm 10\%$ [3]. Furthermore, by setting f_{va} equal to f_v (since measurements were performed at the same location), one can compute the value of K_e consistent with the accurately determined soot volume fraction using the following relationships:

$$f_{va} = \frac{-\ln\left(\frac{I}{I_o}\right)\lambda}{K_e L} = f_v \quad (4)$$

$$K_e = \frac{-\ln\left(\frac{I}{I_o}\right)\lambda}{f_v L} \quad (5)$$

The average value of the dimensionless extinction constant measured for acetylene/air flames in the present study is 8.8 ± 1.5 . These values are somewhat higher than the average value of 8.6 ± 1.5 measured in previous experiments [3]. However, it is believed that the present experiments are more accurate due to direct comparisons that can be made between the light extinction measurements and the sampling measurements.

DISCUSSION

By measuring the dimensionless extinction constant, K_e , we avoid issues related to the refractive index, fractal structure of soot and multiple scattering within the agglomerate in calculating the extinction constant. In this study as well as several others referred to above [1,9,10], the soot volume fraction measurements were made in the post-flame zone. Köylü and Faeth [7] determined that the scattering to absorption ratios for large soot aggregates created in the overfire region of acetylene, ethylene and propane diffusion flames were 41%, 29% and 22%, respectively at $\lambda = 514.5$ nm (The corresponding scattering to extinction ratios are 29%, 22% and 18%). These scattering to extinction ratios are expected to decrease for experiments in which longer wavelength lasers are used (i.e., $\lambda = 632.8$ nm as in the present study). Thus, even for these cases in which scattering is expected to be significant, the estimated total contribution of the scattering to the extinction coefficient at a source wavelength of 632 nm is 23%.

There is also great interest in the soot volume fraction measured within laminar and turbulent flames. The approach that has been widely used is to perform light extinction measurements and then use Eq. 2 with α_{sa} set equal to zero. As discussed above, this leads to an overestimate by a factor of 1.8 (ratio of 8.8/4.9) in the post-flame region. While the soot agglomerates will be smaller in some regions of the flame compared to the

post-flame, they are still large enough that scattering will contribute to the light extinction [6]. For these reasons, the use of $K_e = 8.8 \pm 1.5$ for acetylene measured in the present study is expected to provide more accurate measurement of soot.

Still, more work is needed to better define the degree of scattering in the flame zone. Within the flame, there are also issues to be resolved regarding the refractive index. For example, Habib and Vervisch [11] found differences in the refractive index of soot generated from different fuels (including methane, propane and ethylene). Chang and Charalampopoulos [12] performed experiments using a premixed flame and found variations of the refractive index as a function of height above the burner which suggests influences from the temperature and the soot C/H ratio. For these reasons, experiments are planned to investigate the effects of using different fuels such as ethylene and propane on the measured dimensionless extinction constant of soot.

CONCLUSIONS

By combining gravimetric measurements of the collected soot with soot density measurements using helium pycnometry, accurate values of the soot volume fraction was obtained for rich acetylene/air premixed flames. By calibrating the optical measurements with the gravimetric soot volume fractions, a dimensionless extinction constant, K_e , of 8.8 ± 1.5 was measured. The K_e values measured in this study are recommended as a useful first order estimate for computing soot volume fraction based on light extinction measurements for soot generated from a variety of fuels for both small and large scale flames.

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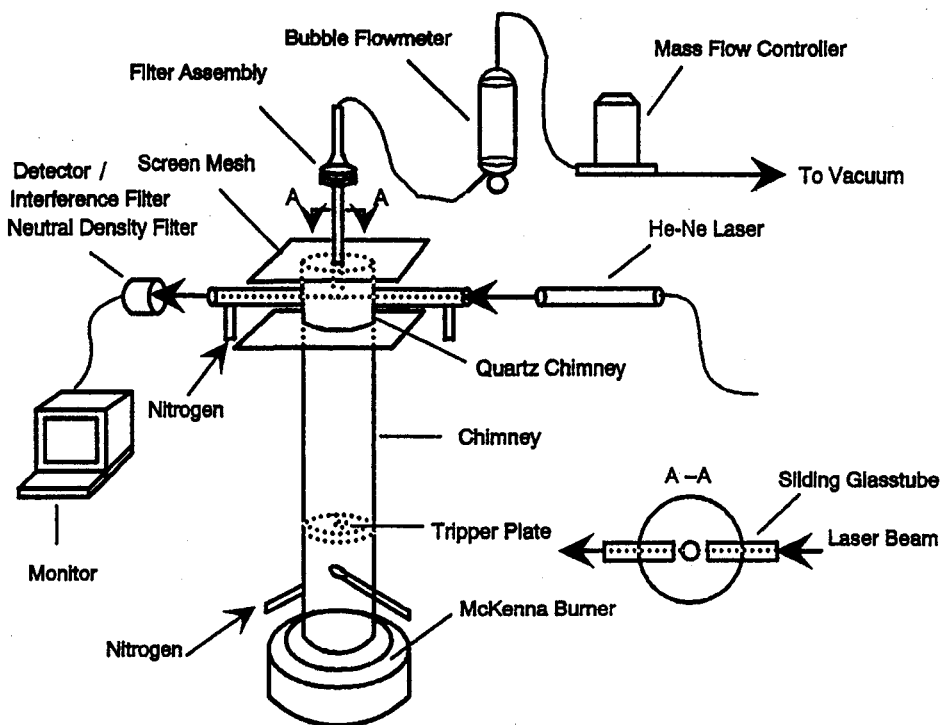


Fig. 1 Schematic of Experimental Apparatus

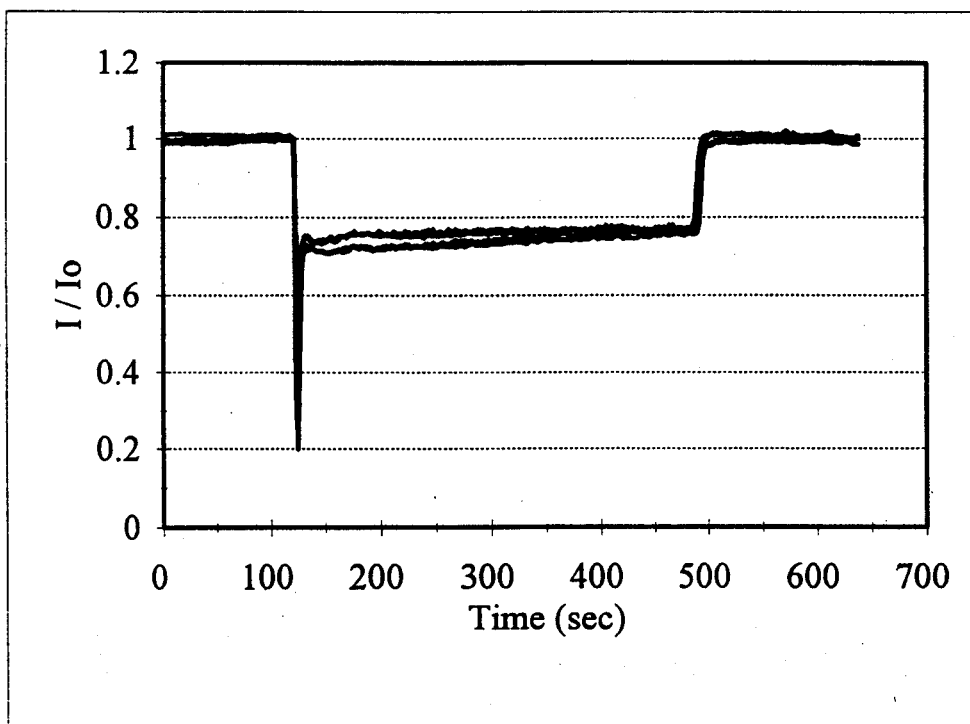


Fig. 2 Light Extinction Measurement for Acetylene/Air Premixed Flame